TUNABLE LASERS

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Tunable radiation in the 0.43μm - 0.52μm spectral region was produced by frequency doubling a tunable dye laser in ZnSe. At wavelengths shorter than the bandgap of ZnSe ($\lambda < 0.46\mu m$), second-harmonic power was limited by crystal absorption, whereas for longer wavelengths the second-harmonic power was as much as 1500 times larger than that predicted for a single-domain crystal. This enhancement is predicted by a simple theory which accounts for the favorable nonlinear optical effects of rotational twins and crystal defects.

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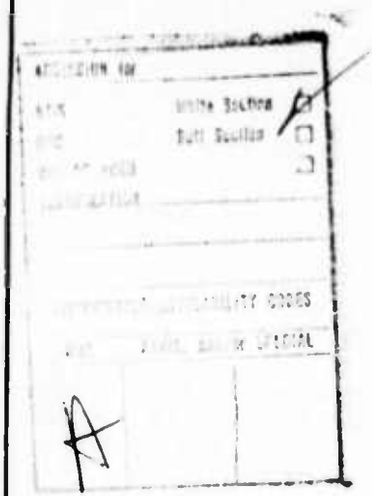
Work has continued on the two photon creation of carriers in ZnSe. It is found that this carrier production method which is dominant at high power levels is very sensitive to wavelength in the 9000 Å region, falling off rapidly to about 1/10th the value of 9200 Å that it had at 9000 Å. These measurements imply that the saturation in infrared difference frequency generation, which we believe to be due to absorption by these free carriers, can be substantially reduced by using longer wavelength pump lasers. A system has been built to check this theory.



TUNABLE LASERS

Semi Annual Technical Report No. 2

For period ending June 30, 1975

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TUNABLE LASERS

ARPA Contract No. N00014-67-A-0204-0092

Technical Report No. 2

For period ending June 30, 1975

Summary

Our search for nonlinear materials suitable for producing tunable infrared power when used in conjunction with a ruby laser and a ruby pumped dye laser has led us to the study of rotational twins in Zinc Selenide. Zinc Selenide, a III-V material which shows the triple attributes of a high nonlinear coefficient, high damage threshold, and large region of optical transparency, unfortunately cannot be phase matched by traditional means as it is not birefringent. We discovered that the sign of the nonlinear polarization is reversed on the two sides of a rotational twin in $\bar{4}3m$ materials and accordingly Zinc Selenide can be effectively phase matched by growing a ZnSe crystal with a regularly spaced, super lattice of twin planes. We did not have at our disposal the capability of producing regularly spaced twins, but by using a piece of ZnSe with randomly spaced twins we were able to show a factor of 250 enhancement over what one could obtain in a difference frequency experiment using a untwinned piece of ZnSe.

We have continued our study of twinned ZnSe and have used it to frequency double a tunable near IR dye laser to the visible. Here an enhancement of over 1000 was observed in comparison to an

untwinned crystal. During this investigation it was also found that an untwinned crystal of ZnSe also gave a bulk effect not unlike that seen with the twinned crystal, although of a much smaller magnitude. This effect was attributed to variations of stoichiometry in the crystal.

The two photon free carrier production coefficient has been studied in the wavelength region around 9000 Å. We feel that the products of free carriers by a two photon absorption of the pump laser beam may be responsible for the observed saturation of the I.R. difference frequency detected at a power level of about 10 MW. The sharp fall off of the coefficient for wavelengths longer than 9000 Å suggests that the power saturation problem could be eliminated by using a pump laser with a longer wavelength. A system has been built to check this theory.

TUNABLE LASERS

ARPA Contract No. N00014-67-A-0204-0092

Semi-Annual Technical Report No. 2

For period ending June 30, 1975

Research Program

1) As a part of our continuing investigation of the use of twin planes for reversing the sign of the nonlinear polarization in Zinc Selenide we have studied frequency doubling in Zinc Selenide in both a twinned and an untwinned crystal. By using a ruby pumped dye laser as a source we were able to study this doubling in a wavelength range for which the harmonic output lies near the short wavelength absorption edge of the material.

2) We have studied the production of carriers in ZnSe by two photon absorption. These carriers may be responsible for the power saturation effect noted in Technical Report #1 of this contract.

3) A new system has been built in which two tunable dye lasers are pumped simultaneously. This system will be of particular value for studying the power saturation effect.

Accomplishments

A) The Effects of Rotational Twin Planes. A paper Enhancement of Second-Harmonic Generation in Zinc Selenide by Crystal Defects by L. O. Hocker and C. F. Dewey, Jr., has been submitted for publication by Applied Physics Letters. This paper reports enhancement

of harmonic generation in a twinned crystal and also in a crystal apparently containing variations from stoichiometry. A preprint of this paper is included as an Appendix to this report.

B) Power Saturation in ZnSe Infrared Difference Frequency Generation. Work has continued on the power saturation effect described in Technical Report #1 of this contract. It was hypothesized in that report that free carriers produced by the intense pump beams partially absorb the infrared radiation produced by the nonlinear interaction of those beams in the crystal. A measurement of the conductivity of the ZnSe crystal while it was subjected to a ruby pulse showed that for low laser powers the conductivity was linearly related to the laser power. At high power levels the relation was seen to be quadratic. These measurements have been repeated but with a tunable dye laser substituted for the ruby.

It was found that for wavelengths shorter than 8800 $\mathring{A}$ the dependence of the conductivity on laser power density was very similar to that found for the ruby laser wavelength. However, for sufficiently long wavelengths the conductivity remained linear in laser power density. This is shown in Figure 1. The absolute power levels shown in figure 1 are only approximate as no power meter was available for this measurement. Relative power levels were determined by a photodiode that had been carefully checked for linearity.

The conductivity of the ZnSe sample can be written as

$$\sigma = a_0 + a_1 P + a_2 P^2 + a_3 P^3 + \ldots \quad .$$

In our measurements the P and P^2 terms dominate, and since a_1 seems to have no substantial wavelength dependence in this range (see fig. 1), the wavelength dependence of the conductivity is entirely attributable to the coefficient a_2. The relative size of this coefficient was measured in the region around 9000 Å and is shown in Figure 2.

The dramatic fall off of this coefficient for wavelengths longer than 9000 Å strongly suggests that by placing both pump lasers at sufficiently long wavelengths the power saturation effect could be reduced or eliminated.

C) Double Dye Laser System. In order to check the hypothesis presented in Technical Report #1 and in part B of this section a system has been built in which two dye lasers are pumped simultaneously by a single ruby laser. The wavelength tuning of these lasers range is such that I.R. wavelengths as short as 10μ can be produced while keeping both dye wavelengths longer than 9200 Å. Time was taken during the construction of this system to substantially upgrade the quality of the optical mounts and components.

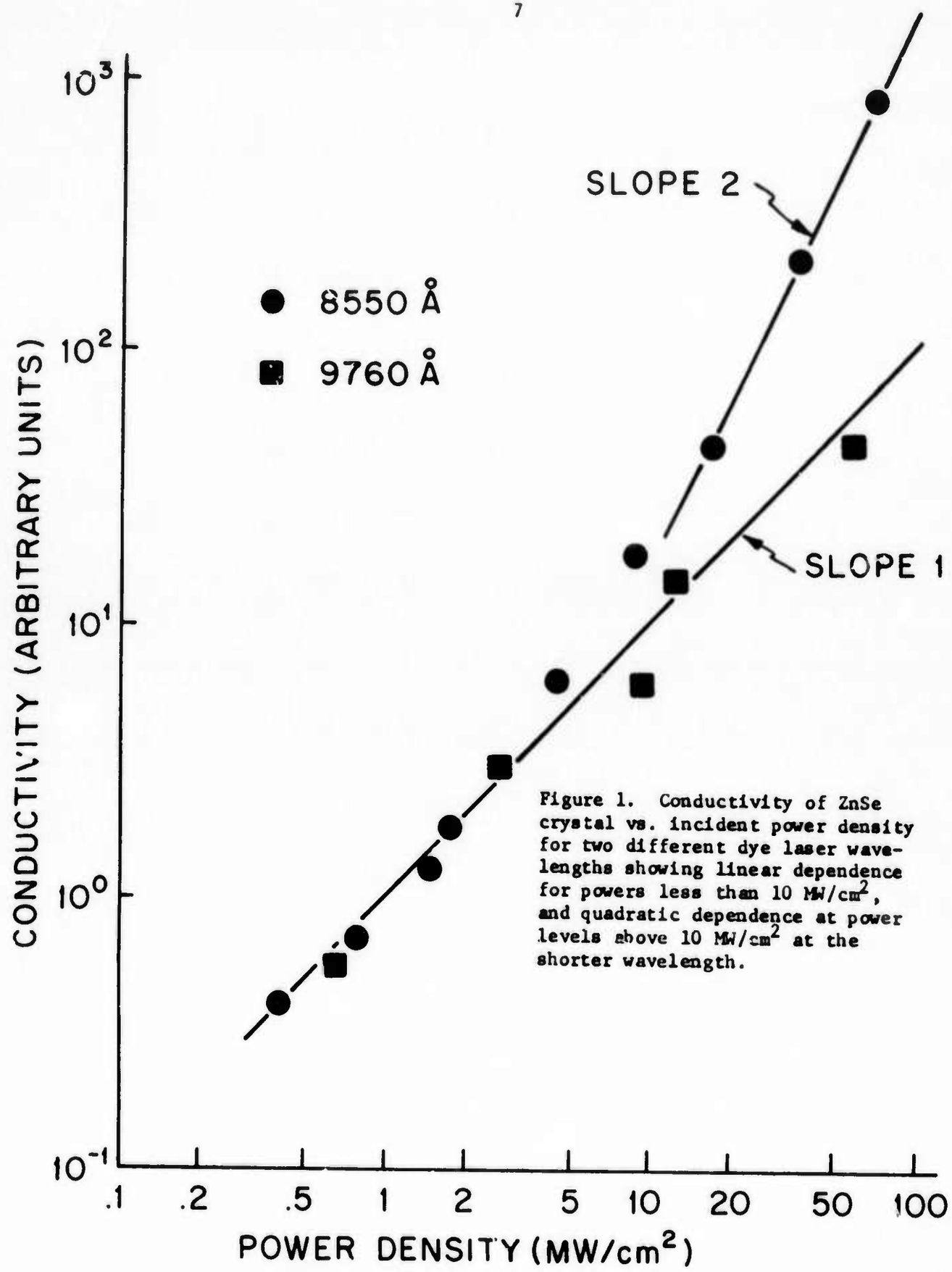

Figure 1. Conductivity of ZnSe crystal vs. incident power density for two different dye laser wavelengths showing linear dependence for powers less than 10 MW/cm^2, and quadratic dependence at power levels above 10 MW/cm^2 at the shorter wavelength.

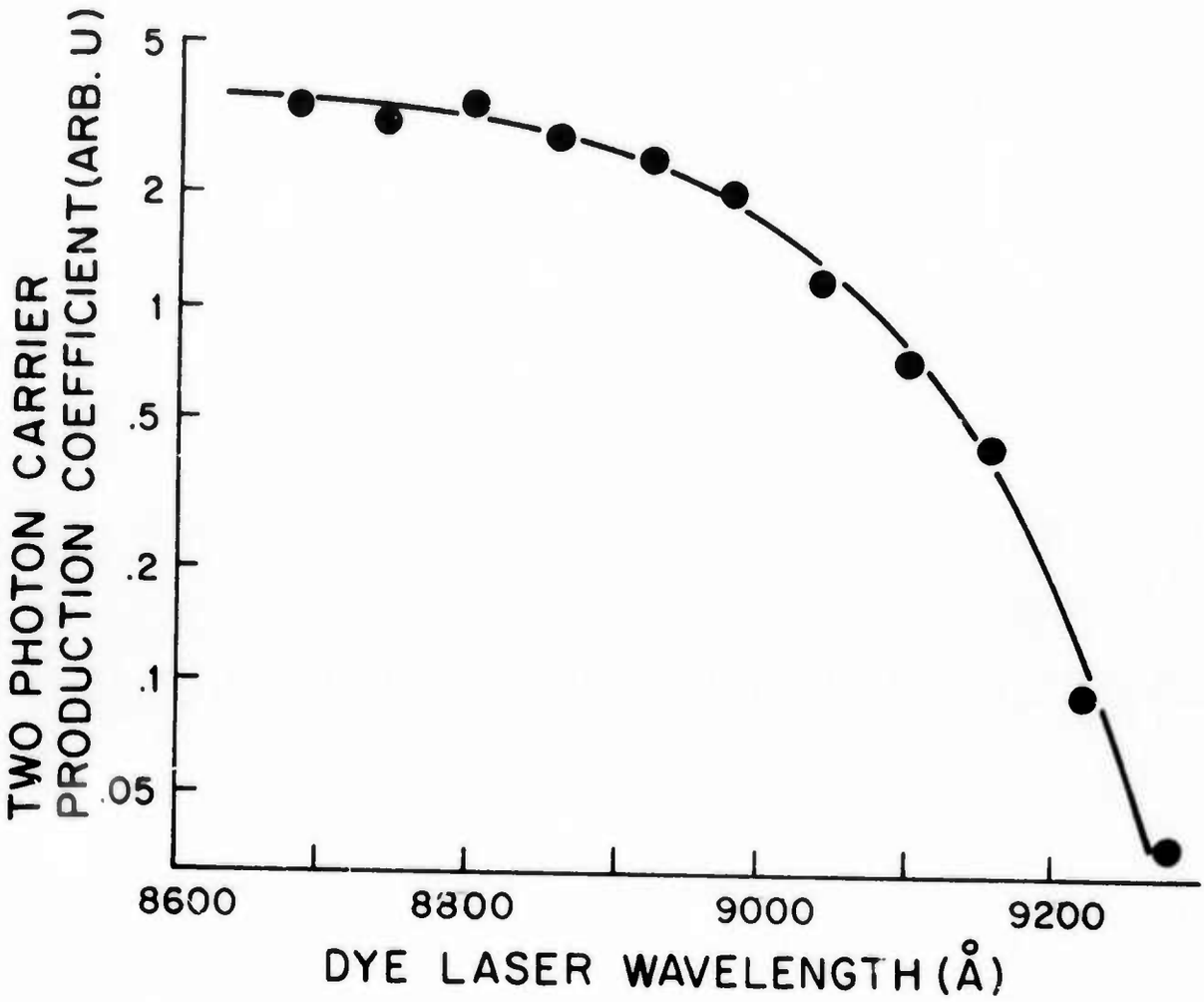

Figure 2. Two photon carrier production coefficient vs. wavelength for ZnSe.

APPENDIX

ENHANCEMENT OF SECOND-HARMONIC GENERATION IN ZINC SELENIDE BY CRYSTAL DEFECTS*

by

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ABSTRACT

Tunable radiation in the 0.43μm - 0.52μm spectral region was produced by frequency doubling a tunable dye laser in ZnSe. At wavelengths shorter than the bandgap of ZnSe ($\lambda < 0.46\mu m$), second-harmonic power was limited by crystal absorption, whereas for longer wavelengths the second-harmonic power was as much as 1500 times larger than that predicted for a single-domain crystal. This enhancement is predicted by a simple theory which accounts for the favorable nonlinear optical effects of rotational twins and crystal defects.

*Research supported by the Advanced Research Projects Agency under Grant N00014-67-A-0204-0092 and the Energy Research and Development Administration under Contract E(29-2)-3597.

Enhancement of Second-Harmonic Generation in Zinc Selenide by Crystal Defects

The attractive optical characteristics of $\bar{4}3m$ materials, which include high damage thresholds, high nonlinear coefficients and large regions of optical transparency, have led several authors to propose schemes that would allow phase-matching of nonlinear optical processes in these crystals[1,2,3,4]. Relatively little experimental work has been performed to demonstrate the efficacy of these techniques. In a recent paper[5], we demonstrated, in Zinc Selenide, that rotational twins are effective in producing enhanced difference-frequency generation.

In this paper we report enhanced second-harmonic generation in Zinc Selenide, both in a twinned crystal and in an untwinned crystal containing random variations in stoichiometry. Second harmonic power was produced at wavelengths in a wide spectral range around the ZnSe band gap. A plot of the power produced can be fit by a very simple model which allows for a bulk second-harmonic production term which is linear in the crystal length as well as terms that one normally considers for an absorbing crystal with a coherence length much shorter than the crystal length. There seems to be no evidence of strong resonant effects near the bad gap such as those seen by Haueisen and Mahr in cryogenicly-cooled CuCl and ZnO[6] or by Faust and Henry in a mixing experiment using I.R. lasers near the reststrahl of GaP.[7]

The laser source used in these experiments was a ruby-pumped dye laser tunable from .72μm to about 1.04μm; the dye laser output had a spectral half-width of about 5 Å and a peak power level of about 3 MW/cm^2 . This tunable pump laser can generate second-harmonic radiation in Zinc Selenide from a region of low absorption above the band gap, down to a region of very strong absorption. Since only that part of the crystal within an absorption length of the surface will contribute to the second harmonic signal, measuring the amount of generated power verses wavelength probes the crystal to different depths. By measuring the second harmonic generated as the dye laser is tuned in the range from 1.04μ to .87μ it is possible to look for bulk harmonic generation from the whole crystal length or from sections as short as a fraction of a micron.

The transmission characteristics of two crystals of ZnSe, one 4mm thick and the other 107μ thick, were measured on a Cary 14 spectrophotometer. The measured absorption coefficient, averaged for the two specimens in the wavelength range 4700 Å to 5200 Å, is shown in Figure 1. Published measurements[8] of the absorption coefficient of thin films of ZnSe show an absorption coefficient of 4.5×10^4 cm^{-1} at 4500 Å with only a small variation between 4200 and 4500 Å. At wavelengths longer than 4650 Å the reported absorption coefficient decreases rapidly. The dashed line is an extrapolation in the spectral region not covered by either our data or the data of Zhalkevich[8].

The wavelength dependence of the second-harmonic power was measured for two ZnSe Crystals. This dependence normalized to the square of the dye laser output is plotted in Figure 2. The large random variations of the observed signal are attributed to variations in the dye laser output mode pattern. (Normalizing to the intensity of the second harmonic emitted by a non-phase-matched ($\Delta K \neq 0$) crystal of ADP as done by Haueisen and Mahr[6] would have eliminated this variation.) For these measurements, the dye laser beam was propagated along the 111 direction of the crystal.

The squares in Figure 2 are the points taken for a ZnSe crystal with many twin planes. In a previous experiment[5] this crystal had been used to produce the difference frequency between a ruby laser and a tunable dye laser. An enhancement of about a factor of 250 over that which could be produced by one coherence length of untwinned material was observed in that measurement.

The circles refer to data taken for another crystal which showed no evidence of twins when examined under a microscope using unpolarized light (see Fig. 3a). Examination of the crystal under a microscope using crossed polarizers showed strong evidence of birefringence. Figures 3b and 3c, respectively, are pictures of the crystal taken with the surface parallel and perpendicular to the 111 axis of the crystal using the crossed polarizers. These pictures suggest that variations of stoichiometry during the crystal growth are responsible for the observed birefringence as the 111 direction is the growth direction and striations are observed perpendicular to the direction of growth (Fig. 3b).

It is possible to calculate an approximate expression for the second-harmonic power radiated from a randomly-twinned absoring crystal containing N twins. We assume that the twins, although randomly spaced, are roughly evenly distributed along the length of the crystal and are all perpendicular to the 111 direction. We also assume that Miller's rule[9] is applicable so the wavelength dependence of the nonlinear coefficient comes in only through the wavelength dependence of the index of refraction, n. Since the two sides of our crystal were not plane parallel, the oscillatory terms giving Maker fringes are averaged out. Under these circumstances, the second-harmonic power, $P_{2\omega}$, is given by

$$P_{2\omega} = G \cdot P^2(\omega) \frac{(n_2^2 - 1)(n_1^2 - 1)^2}{n_1^2 n_2 \lambda_2^2} \frac{1 + e^{-\alpha_2 L} + \frac{2N}{\alpha_2}[1 - e^{-\alpha_2 L}]}{(\Delta K^2 + \alpha_2^2/4)} \quad (1)$$

Here G is a constant independent of wavelength, $P(\omega)$ is the laser intensity, $\Delta K = \frac{4\pi}{\lambda_1}(n_2 - n_1)$, α is the absorption coefficient, L is the length of the crystal, λ is the wavelength of the light within the crystal, and the subscripts 1 and 2 refer to the fundamental and harmonic waves respectively. We have assumed that the crystal absorption at the fundamental wavelength is negligible, and that the variation of input and output reflection losses with wavelength are unimportant.

We chose the index of refraction of ZnSe to be given by

$$n^2 = 3.855 + 2.045\lambda^2/(\lambda^2 - .109) \quad . \quad (2)$$

15

The coefficients of Eq. (2) are the averages of those found by Marple[10] to give best fits to for his two ZnSe prisms. (Some uncertainty exists in using the index values of Eq. 2 for short wavelengths, as Marple's measurements extend only to .475μ.)

Eqs. (1) and (2) may be used to describe the experimental results obtained on the two crystals we tested. There are a total three unknown parameters for the two sets of data of Fig. 2. One appears in the term $GP^2(\omega)$; inasmuch as relative rather than absolute powers were measured, the term $GP^2(\omega)$ contains a multiplicative constant which is the same for the two crystals. The remaining free parameters are the number of "twin planes" in each of the crystals. The best fit between the experimental points and the theory was found when the value of 1600 twins was chosen for the "twinned" crystal and 18 twins for the "untwinned" crystal. Also shown on figure 2 is the curve one would expect for a crystal with no twins. The curve with no twins, and the curve with 18 twins coincide for wavelengths less than .46μ.

The value of 1600 twins for the twinned crystal is not unreasonable although it is larger than expected. Using a microscope we were able to detect about 300 twin planes with separations greater than about 2μ. The coherence length for the harmonic generation process is about 1 micron for a 1μ input wavelength. Since twins spaced even less than half the coherence length can contribute significantly, it is clear that observations with a light microscope will only provide a lower limit to the number of effective twins.

The discrepancy between the experimental points and the theoretical curve for fundamental wavelengths shorter than .92μ could be caused by the approximation that the twin planes are evenly distributed throughout the crystal. For long absorption lengths the power generated will be only a weak function of the plane positions. However for absorption lengths comparable to the distance from the output face of the crystal to the first few twins in from that face, the exact position of these few twin planes becomes important. One twin plane within an absorption length of the surface would be more than sufficient to explain the observed deviations.

The results for the "untwinned" crystal require quite a different explanation. A careful study of the surface of the crystal showed no evidence of twins. If a surface is perpendicular to the twin planes of a twinned crystal, this surface will show different light reflecting characteristics on the two sides of the twin plane. As one traverses the crystal surface in the 111 direction, the surface the reflection characteristics change back and forth as each twin plane is crossed. An even number of twins spaced more closely than the resolution of the system would appear as a line with the same reflection characteristics on either side. Were these twins spaced very much closer than the resolution of the instrument, even this line would disappear. It is, however, unreasonable to expect that twins appeared in this crystal in very closely spaced groups of even numbers, and one is left with the conclusion that the enhanced harmonic generation which the "untwinned" crystal displays must be due to an effect other than twinning.

An examination of the untwinned crystal under a microscope using

crossed polarizers showed striations perpendicular to the 111 axis (Fig. 3c). These striations could be the result of strains[11] resulting from slight variations from stoichiometry during the crystal growth[12]. Stoichiometry variations would produce index of refraction variations, and hence, according to Miller's rule, modulation of the nonlinear coefficient. Refractive index modulation and/or nonlinear coefficeint modulation[1-4] would give rise to an enhancement of second harmonic generation analogous to the enhancement observed with rotational twins. Both of these effects have been explored theoretically by Tang and Bey[2]. For either effect to explain the bulk effect equivalent to 18 twin planes, the appropriate Fourier component of the index of refraction would have to have an amplitude, Δn, of about 0.001. Since the enhancement is observed to take place over a very large bandwidth, the total modulation must be quite large. Much greater second-harmonic conversion efficiencies could be achieved if this modulation amplitude could be restricted to one Fourier component; however, as Tang and Bey have pointed out, good conversion efficiencies can only be reached for very high index modulations.

Local strain-induced birefringence caused by random variations of stoichiometry could also be the source of the bulk term without any assistance from a nonlinear coefficient modulation. This would appear as a coherence length modulation in a manner similar to that of refractive index modulation. To our knowledge this effect has not been studied theoretically.

It is interesting that this simple model seems to describe the wavelength dependence of the measured conversion for both samples. There are no strongly resonant terms in the nonlinear coefficient as described by the Miller's rule approximation.

Although it is unlikely that index-modulated crystals will prove to be competitive materials for frequency doubling into the visible, the effects of index modulation cannot be overlooked for two reasons. First, in systems for obtaining tunable infrared by mixing near-infrared sources, this process may be practical considering the relatively poor conversion efficiencies available using any system. The high nonlinear coefficients, damage thresholds and large regions of transparency of materials such as ZnSe, ZnS and ZnTe coupled with this mechanism for making them phase matchable could produce larger infrared difference frequency powers than presently obtainable from conventional crystals. And second, unintentional index modulation may be a significant source of error in experiments for determining the nonlinear coefficient of crystals. This would be of particular importance for measurements of materials with band gaps in the infrared, because the uniformity of these materials is difficult to determine. Phase-matchable crystals could also exhibit such modulations, leading to unexpectedly short coherence lengths.

Crystal growth processes[13] are not understood well enough at the present time to make possible the manufacture of crystals with controlled rotational twins or controlled variations in stoichiometry and strain. The incentive for producing periodically-spaced rotational twins is clear; coherent rather than random addition of the nonlinear polarization of each domain would produce nonlinear effects proportional to the square of the number of domains, rather than the linear variation we have observed.

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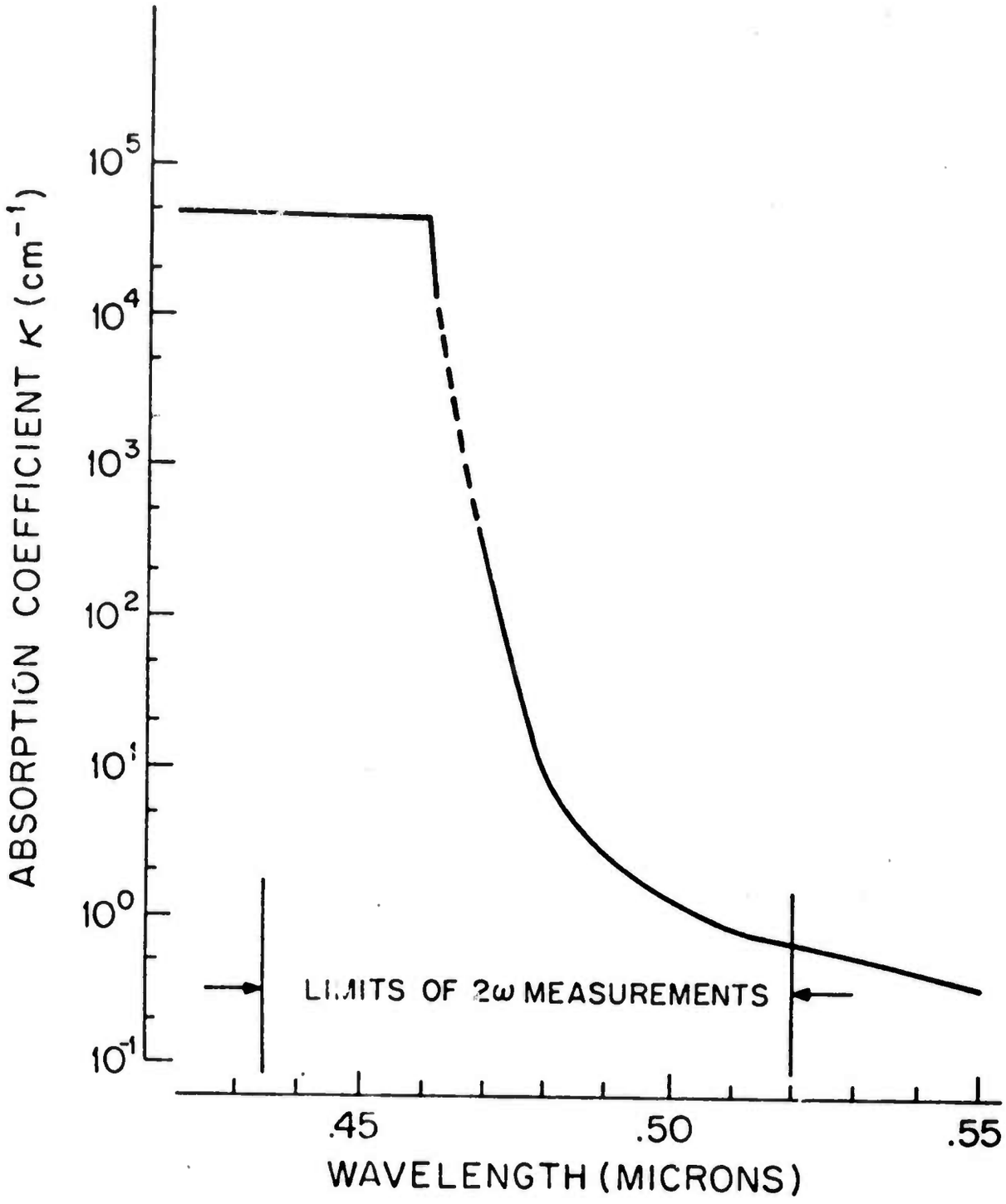
Figure Captions

Figure 1. Absorption coefficient of ZnSe verses wavelength. Values for $\lambda < .46\mu m$ are from Ref. 6; for $\lambda < .47\mu m$ measurements are from this work. Dashed part of curve is interpolated.

Figure 2. Second harmonic generation coefficient measured for twinned crystal (squares) and "untwinned" crystal (circles). The curves are the expected coefficient for a ZnSe crystal containing 1600 twins (top curve), 18 twins (middle curve) and no twins (bottom curve).

Figure 3. ZnSe crystal viewed under a microscope: a) side view, no polarizer; b) side view, crossed polarizers; c) face view crossed polarizers.

Fig 1


ABSORPTION COEFFICIENT K (cm^{-1})
10^5
10^4
10^3
10^2
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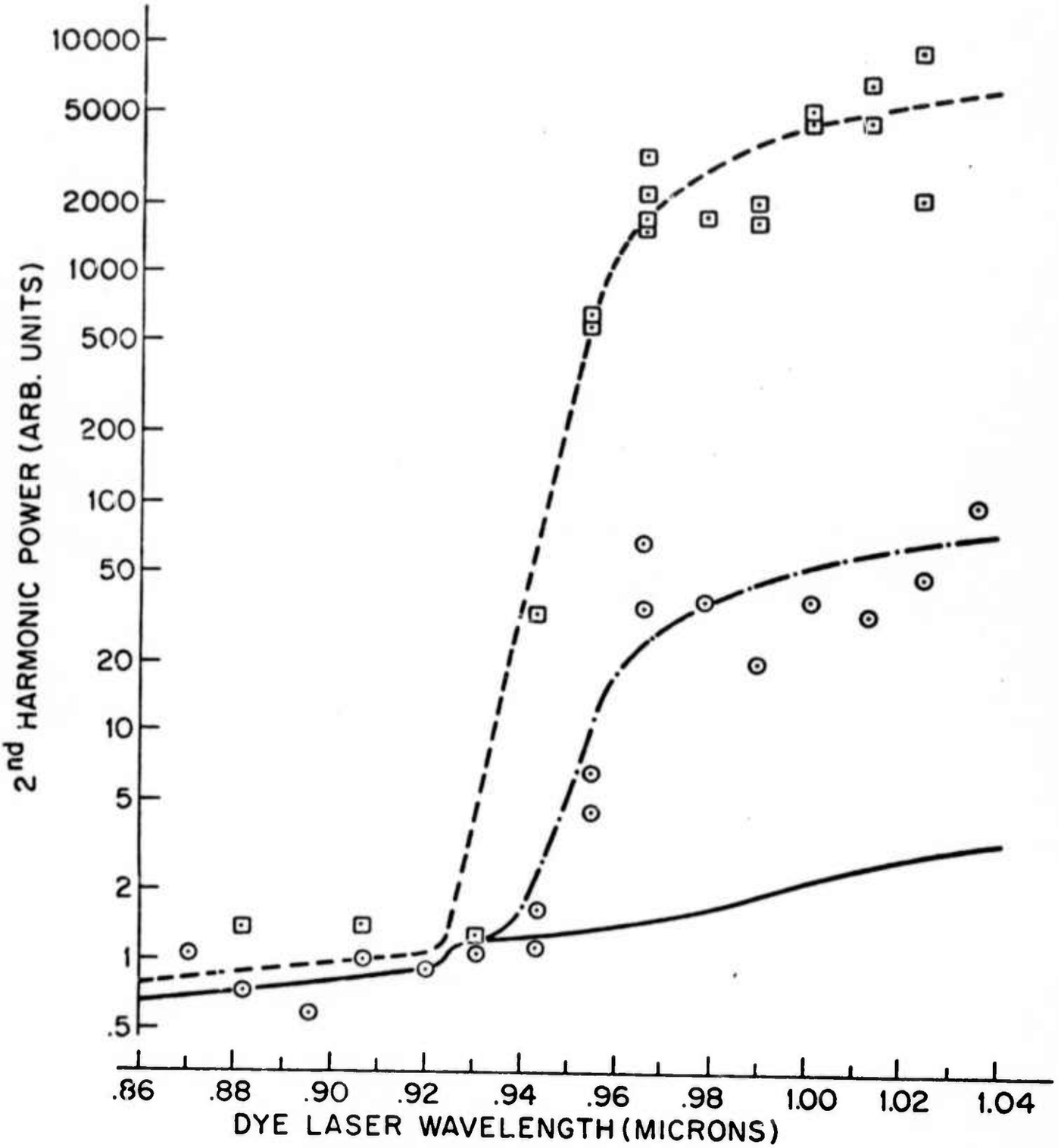
10^0
10^{-1}
LIMITS OF 2ω MEASUREMENTS
.45
.50
.55
WAVELENGTH (MICRONS)

Fig 2


10000
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2nd HARMONIC POWER (ARB. UNITS)
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DYE LASER WAVELENGTH (MICRONS)

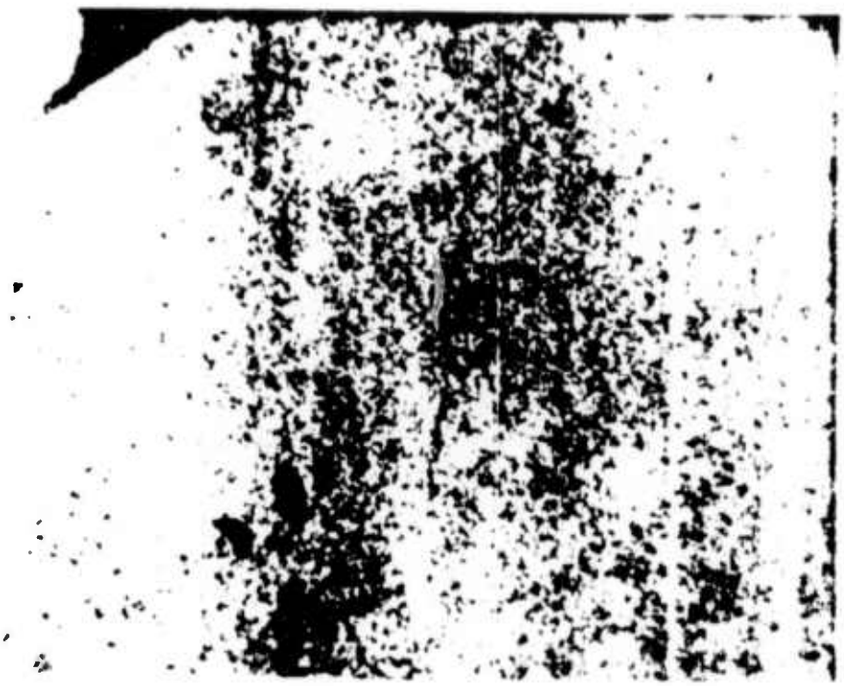

FIG 3A

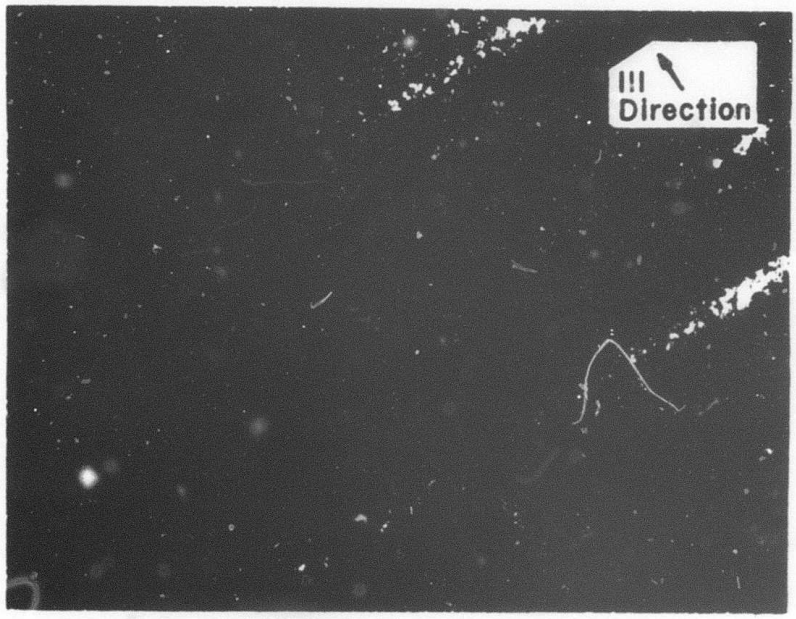

Fig 3b

Fig 3C

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